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APPROXIMATE SCALING LAWS OF HOMOGENEOUS CONDENSATION IN EXPANDING SUPERSONIC GAS FLOWS

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Due to the extreme complexity of the process of nucleation and growth of condensed-phase particles, the problem of gas flow with condensation cannot be solved analytically even with simple unidimensional flow models. The author of [1] used the example of flows of a condensing gas in supersonic nozzles and analyzed the corresponding dimensionless equations to show that exact modeling is also impossible. At the same time, the existence of different empirically established correlations connected with condensation in supersonic flows suggests that it is possible to find approximate similarity. There has been a whole range of studies devoted to establishing approximate scaling laws for condensation in expanding supersonic gas flows. The difference in the approach to the solution of the given problem and in the formulations and the generality of the resulting laws make it incumbent to conduct a comparative analysis of these investigations.

The study [2] was the first investigation to sufficiently thoroughly establish the scaling conditions, using as an example the approximate solution of the problem of condensation in a cloud of a vaporized substance during spherical dispersion into a cavity. Condensation kinetics was described by using the classical Frenkel–Zeldovich formula for the rate of formation of critical nuclei as a function of the degree of supercooling ϑ: $I = c\exp(-b/\vartheta^2)$. Here, $\vartheta = (T_p - T)/T_p$ (T_p is the temperature of vapor saturated at the given density). The extremely heavy dependence of the rate of nuclei formation on the degree of supercooling leads to a situation whereby most of the condensation centers ν are formed on a very small section of the expansion stage corresponding to maximum supercooling ϑ_m:

$$\nu = \int_{t_s}^{\infty} I\,dt' \approx I(\vartheta_m)\,\Delta t_m$$

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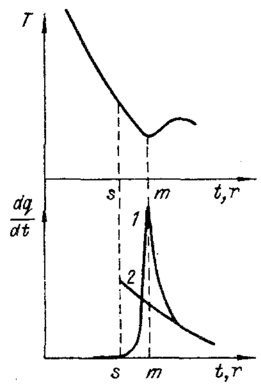

Fig. 1

(t_s is the time corresponding to the moment of attainment of the state of saturation). Due to the abrupt change in nucleation rate, the rate of the change in the fraction of condensate dq/dt also exhibits a heavy dependence in the region of maximum supercooling. Figure 1 schematically depicts the dependence of the gas temperature and condensation rate on the expansion coordinate (the time t or distance r) (see, e.g., [3]). Here, the point s is the moment of attainment of the saturated state. The minimum of the gas-temperature distribution, the supercooling maximum, and the maximum of the rate of nonequilibrium condensation (curve 1) lie within a small region around point m. The figure also shows the curve for the rate of equilibrium condensation (curve 2). It is equal to zero at the moment of saturation, undergoes a discontinuity at the point s – where it is maximal from the right – and then decreases with further expansion of the gas. The narrowness of the distribution function of dq/dt near point m makes it possible to determine the position of the latter from an equation of the form

$$(dq/dt)_\xi = Q(\xi), \qquad (1)$$

in which some arbitrariness is permitted in the choice of the function Q. The quantity ξ is a certain point from the neighborhood of m. As the function Q, we can choose the equilibrium condensation rate [4] or even its higher value at the moment of saturation. The function $y = (u/L)(du/dt)$ was chosen as such in [2]. As follows from the energy equation

$$c_p dT - L dq + u du = 0 \qquad (2)$$

(c_p is the isobaric heat capacity; L, heat of phase transformation; u, velocity of the gas flow), this function corresponds to the condensation rate at the point where $dT/dt = 0$, i.e., near ϑ_m. However, the arbitrariness lies in the fact that we used the value of y at the saturation point to solve Eq. (1). Approximate solution of (1) leads to a transcendental equation for ϑ_m [2]: $1/I(\vartheta_m) = F(\vartheta_m, T_s, t_s, p_s)$ (p_s is the pressure at the moment of saturation). Thanks to the exponential character of $I(\vartheta)$, the amount of maximum supercooling remains very stable. Knowledge of ϑ_m makes it possible to determine the total number of condensation centers (from calculations performed for one molecule): $\nu \approx \Phi(\vartheta_m, T_s)(t_s p_s)^{-3}$. A change in the initial conditions is accompanied by a slight change in T_s due to the exponential character of the elasticity curve of the vapor. Thus, the condition for maintenance of the number of condensation centers is $t_s p_s = \text{const}$. It was shown in [2] that this is equivalent to the requirement of conservation of the number of collisions that a vapor molecule undergoes from the moment of saturation to infinity.

The above-formulated scaling condition can be made more restrictive by taking into consideration that the gas expands nearly isentropically up to the moment of saturation, i.e., the condition can be modified by requiring an isentropic change in the initial conditions. Then the scaling condition reduces to the requirement of retention of the scaling time $t_s = \text{const}$.

The requirement of retaining the time scale with an isentropic change in the initial conditions can be satisfied if we do not directly integrate the equations of the condensation kinetics (and gasdynamics). To make this possible, it is sufficient to compare the growth of clusters at identical temperatures in an elementary temperature range $(T, T - dT)$, assuming that the condensation mechanism is determined mainly by first- and second-order reactions (spontaneous vaporization, binary collision of clusters with monomers) and that the flow is isentropic before condensation begins [5]. Examination of the kinetic conditions which govern cluster growth makes it possible to obtain modeling conditions for a constant nozzle geometry, as well as a constant stangation temperature. For axisymmetric flows, the dependences of the time interval dt and the number of collisions dz on the initial conditions are determined by the following expressions:

$$dt \sim -dTd_{eq}T_0^{(2-\gamma)/(2\gamma-2)}; \tag{3}$$

$$dz \sim -dTd_{eq}p_0T_0^{(2-3\gamma)/(2\gamma-2)}. \tag{4}$$

Here, γ is the ratio of the heat capacities; p_0 and T_0 are the stagntion temperature and pressure; d_{eq} is the "equivalent" nozzle diameter, introduced in [5]. With an increase in T_0 and a constant nozzle geometry, stagnation pressure must increase in accordance with the following law in order to retain dz:

$$p_0 \sim T_0^{(3\gamma-2)/(2\gamma-2)}. \tag{5}$$

However, in this case, there will be no compensation for spontaneous vaporization. The contribution of the latter increases as a result of an increase in the time interval. If we assume that, on the average, the increase in dz leads to growth of the cluster, then spontaneous vaporization may be compensated for more rapidly than in (5) by an increase in p_0: $p_0 \sim T_0{}^r$ $[r > (3\gamma - 2)/(2\gamma - 2)]$. If p_0 increases in accordance with the more rapid isentropic law, i.e., $p_0 \sim T_0{}^{2\gamma/(2\gamma-2)}$, then the only consequence of an increase in T_0 will be an increase in dt — such slowing of the flow leads to growth of the cluster. This means that, with a constant nozzle geometry, the scaling law has the form

$$p_0T_0^{-r} = c\,(d_{eq}), \tag{6}$$

where $2\gamma/(2\gamma - 2) > r > (3\gamma - 2)/(2\gamma - 2)$.

With a constant temperature in the precombustion chamber, d_{eq} should decrease in order to maintain dz when p_0 increases. Any reduction in d_{eq} leads to a decrease in dt and, accordingly, to a decrease in the contribution of spontaneous vaporization. Here, a change in d_{eq} at a rate slower than $d_{eq} \sim 1/p_0$ also leads to an increase in dz. Thus, with an increase in p_0, it is necessary to have d_{eq} decrease more rapidly than $p_0{}^{-1}$, specifically, $d_{eq} \sim p_0{}^{-\beta}$, where $\beta > 1$. Summing up, the scaling law for a constant temperature T_0 is represented in the form

$$p_0d_{eq}^q = c\,(T_0) \quad (0<q<1). \tag{7}$$

As indicated above, the only consequence of an isentropic change in the initial conditions with d_{eq} = const is a change in the time interval. The dependence of the latter on just two parameters (d_{eq} and T_0) makes it possible to keep the interval dt constant through a corresponding change in d_{eq}. Thus, it is possible to formulate conditions for "exact" modeling with a simultaneous change in all three parameters [6]:

$$p_0 = c_1T_0^{2\gamma/(2\gamma-2)}, \quad d_{eq} = c_2T_0^{(\gamma-2)/(2\gamma-2)}. \tag{8}$$

The same result was obtained in [7] using the example of the approximate solution of a problem which entailed finding the maximum supercooling in a hypersonic flow in nozzles with rectilinear generatrices. The scaling conditions formulated in this study for a conical nozzle have the form

$$S_0 = \text{const}, \quad \varphi \equiv (r_*/\text{tg}\,\alpha)\,T_0^{(2-\gamma)/(2\gamma-2)} = \text{const}, \tag{9}$$

where S_0 is the entropy of the flow; r_* is the characteristic dimension of the critical cross section; α is the angle between the generatrix and the nozzle axis. For a conical nozzle, the value of the time interval dt corresponding to the expansion section (T, T − dt) is connected in a hypersonic approximation with the function φ by the relation $dt = dT\varphi f(T)/\sqrt{2c_p}$. In this relation, f(T) is a certain function of temperature. Thus, the condition φ = const is equivalent to dt = const for the prescribed time interval (T, T − dt), which means that laws (8) and (9) are identical.

Scaling laws (6)-(8) were obtained from examining the kinetic conditions of cluster growth on an elementary temperature interval, without analyzing the expansion process as a whole. This approach does not require knowledge of the temperature dependence of the reaction cross sections, but it also does not consider changes in the total number of effective collisions due to a change in the temperature T_s at which saturation is achieved. Nevertheless, allowance for this factor makes it possible to obtain scaling laws in the same form on the basis of the condition of retention of the number of binary collisions beyond the phase transition curve. Also, it is necessary to restrict the analysis to binary collisions in order to obtain these laws. This approach was used in [8] to obtain condition (7). It is not hard to show that other scaling laws (6)-(8) can be found in a similar manner. For example, let us take condition (6). With a constant nozzle geometry and a fixed temperature in the precombustion chamber, the number of binary collisions z after the saturation point, due to the dependence of its position on p_0, $p_0^{\varkappa}$ $(\varkappa > 1)$: $z \sim d_{eq}\, p_0^{\varkappa} T_0^{(2-3\gamma)/(2\gamma-2)}$. From here, the condition z = const gives (with d_{eq} = const), $p_0 T_0^{(2-3\gamma)/\varkappa 2(\gamma-1)} =$ const or $p_0 T_0^{-r}$ = const. Meanwhile $r > (3\gamma - 2)/(2\gamma - 2)$. To obtain the upper bound for r and Eq. (8), the initial conditions should be subjected to an isentropic change. Since this does not change the limits of integration to calculate the number of collisions (the position of the saturation point remains the same), the condition z = const will be equivalent to dz = const — the problem is again reduced to analysis of an elementary expansion section.

The scaling laws examined above are not so much a fruit of theory as they are a fact of empirical observation. Numerous experiments using molecular-beam technology have shown that laws (6)-(8) generalize the conditions of formation of clusters of a prescribed mean size in the long-range field of a jet [5, 9], as well as the conditions under which monomer velocity begins to increase, the dimer signal becomes maximal [10], and the intensity of the molecular beam becomes minimal [8]. Similar laws have been established for the spatial position of the beginning of condensation — determined from measurements of Rayleigh scattering — and for the behavior of scattering intensity in the condensation zone [11]. Our experiments have confirmed the presence of boundaries for the exponents r and q. The range established for q is much narrower than the range $0 \leq q \leq 1$. Within the experimental error, these quantities turned out to be constant for the given species of gas and change within the range $2.375 > r > 2.25$ for monatomic gases (depending on the gas) and within the range $3.125 > r > 2.75$ for diatomic gases. Here, $0.5 < q < 1$ [6].

The accuracy of the experiment proved to be adequate to detect a variation of r in the case with CO_2 [5], due to the nonideality of this gas. The model of equilibrium spherical flow was used in [12] to modify law (6), which determines the conditions of condensation of CO_2.

The narrowness of the range for r and q and their near-constancy for monatomic and diatomic gases makes it possible to combine relations (6) and (7) into the scaling law

$$p_0 T_0^{-r} d_{eq}^{q} = c_N, \tag{10}$$

which in logarithmic coordinates is the equation of a plane; it allows and includes an isentropic change in the initial parameters p_0 and T_0. Here, compatibility conditions (10) and (8) lead to the following relations: $q(2 - \gamma)/(2\gamma - 2) = 2\gamma/(2\gamma - 2) - r$, $c_2 = (c_N/c_1)^{1/q}$.

Thus, we have significantly reduced the number of experiments needed to establish specific correlations when studying condensation.

Until now, when examining scaling laws in condensing jets, most investigators have assumed that the change in the state of the gas during expansion up to the saturation point occurs isentropically. The studies [13-15] developed an analytical approach to which an arbitrary law of change in the state of the gas $p = f(T)$ is adopted. Here, a separate analysis is made of condensation at the initial stage — when the effect on gasdynamics can be ignored — and at the stage of developed condensation. An example of the criterion which allows these stages to be distinguished from one another is the ratio of the heat of condensation to the enthalpy of the mixture:

$$Lq_m/h_m = c_m. \tag{11}$$

If the temperature of the gas is taken as an independent variable, then the kinetic equation can be represented in the form

$$dq/dT = \omega(p, T, q)\dot{T}^{-1}, \tag{12}$$

where the dot denotes differentiation with respect to time. The rate of cooling $\dot{T}$ changes in the active condensation region much more slowly than the function ω. Thus, such a representation simplifies the analysis.

During the initial stage of the process, $\omega \approx \omega(T)$, and the kinetic and gasdynamic problems are separated. Then for q at the point m we have

$$q_m = \int_{T_s}^{T_m} \dot{T}^{-1}\omega(T)\,dT,$$

or, taking $\dot{T} \approx \dot{T}_s$ on the integration section,

$$q_m = \dot{T}_s^{-1}\Phi(T_m, T_s). \tag{13}$$

Since $h \approx h(T)$ during the initial stage, it follows from (11) and (13) that $T_m = T_m(c_m, T_s, \dot{T}_s)$. Similarly, it can be shown that the mean size of the clusters is also a function of these parameters. This means that with a specified law of expansion in the initial stage $p = f(T)$ and a chosen criterion c_m, the governing parameters are the temperature at the saturation point T_s and the cooling rate $\dot{T}_s$.

At the stage of developed condensation, it is necessary to simultaneously solve the gasdynamic and kinetic equations. Assuming that $h = c_pT - Lq$ and again choosing the temperature as an independent variable, we write the energy equation in the form

$$d\ln p/d\ln T = (c_p - L\,dq/dT)/R_\mu(1 - q). \tag{14}$$

If in regard to the function $\dot{T}$ we can again a priori represent $\dot{T} = \dot{T}(p, T, q)$, without time derivatives, then the simultaneous solution of (12) and (14) with the prescribed initial conditions $p(T_m) = p_m$, $q(T_m) = q_m$ determines the condensation process – p(T), q(T), etc. Thus, the governing parameters are T_s and $\dot{T}_s$ in this case as well. The set of initial states p_0, T_0 satisfying the condition T_s = const consists of those states belonging to the single expansion curve $p = f(T)$. From this, it is not hard to see that the scaling conditions T_s = const, $\dot{T}_s$ = const nearly coincide with the laws obtained earlier for an isentropic law of expansion at the initial stage. It was shown above for such flows that an isentropic change in the initial states does not ensure $\dot{T}$ = idem. However, on the one hand, it can be seen from (3) that the dependence of the cooling rate on T_0 is relatively slight and, on the other hand, so is the effect of the rate itself $\dot{T}$ on the solution of a flow problem with condensation. Thus, the calculations performed in [4] for nitrogen showed that in the case of an appreciable difference in the results of determination of the beginning of condensation, the rates of expansion should differ by a factor greater than 20. In [15], results presented from calculations performed for flows of water vapor confirm these conclusions.

Let us discuss the scaling conditions for the homogeneous condensation of different gases. This question was examined in [16, 17]. The main conclusion reached in these studies is essentially that for thermodynamically similar gases, the laws of the respective states can be extended to flows in jets with condensation. This means that for such gases the scaling conditions formulated above – being reducible to dimensionless form – are universal in character. The validity of the scaling conditions for methyl acetylene C_3H_4 and chloroform $CHCl_3$ was confirmed by calculation [17]. Here, the temperature and pressure at the critical point were taken as the reference state parameters of the gas. The fact that the inert gases belong to a single class of "similar" substances was demonstrated experimentally in [5, 18] for the mean cluster size and in [19] for the mass fraction of condensate. The authors of these studies converted variables to dimensionless form by using parameters of the Lennard–Jones potential.

Let us examine yet one more experimental study, also devoted to the condensation of inert gases and generalization of the results in dimensionless form [20]. The authors studied condensation in the flow of Ar, Kr, and Xe in a mixture with He in small and very long Laval nozzles. This is an instance of a fairly complicated flow requiring for analysis consideration of the effect of viscosity and differences in the molecular weights of the components of the mixture. Naturally, efforts to generalize results on condensation in such flows – and even more so for different gases – will enjoy more limited success.

The generalization of the beginning of condensation performed by the authors of [20] with the use of parameters of the molecular interaction potential cannot be considered correct, and they improperly concluded that the use of the corrected parameter $\tilde{p}_0 = p_0(\varepsilon/\sigma^3)$

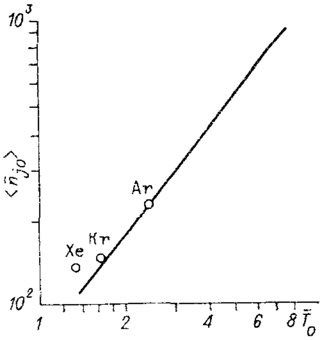

Fig. 2

TABLE 1

Gas	$\bar{T}_0$	μ, kg/kmole	$\bar{m}$	$(\bar{m}/m_j)^{0,5}$	$1/\sigma$	$p_{0j}\sigma^3$	const from (18)	$\langle \bar{n}_{j0} \rangle$
Ar	2,46	40	6,16	0,39	0,29	236,9	8,57	236,9
Kr	1,72	83,8	8,79	0,32	0,28	196	8,86	154,2
Xe	1,33	131,3	11,6	0,297	0,24	234	11,69	147,5

for generalization "does not work." Their principal mistake was that they attempted to compare results obtained with significantly different corrected temperatures $\bar{T}_0 = T_0/(\varepsilon/k)$. Shown below is an example of the more proper use of the principle of corresponding states to analyze data on the beginning of condensation.

In accordance with the principle of corresponding jets, the conditions of cluster growth are identical if the following corrected quantities are also identical:

$$\bar{p}_0 \equiv p_0/(\varepsilon/\sigma^3) = \text{const}, \ \bar{T}_0 \equiv T_0/(\varepsilon/k) = \text{const}, \\ \bar{d}_{eq} \equiv d_{eq}/\sigma = \text{const}, \ \gamma = \text{const}. \tag{15}$$

The study [20] presented measurements of the intensity of a molecular beam as a function of pressure p_0 at constant values of temperature T_0 and the molar fraction x of the condensing gas in the mixture and a constant nozzle geometry. It is clear that the points of the beginning of condensation $\bar{p}_{0j}$ determined from this data for different gases cannot be compared directly, since the remaining corrected parameters are different. These results can be checked for satisfaction of the scaling laws.

For a flow of a mixture of monatomic gases, the expressions for the time interval dt and the number of collisions dz in the spherical flow model have the form

$$dt \sim -dT f_1(T) f_2(\gamma) r_* \bar{m}^{0,5} T_0^{(2-\gamma)/2(\gamma-1)}; \tag{16}$$

$$dz \sim -(dT/T) r_* f_2(\gamma) n_{j0} (\bar{m}/m_j)^{0,5} (T_0/T)^{\gamma/2(1-\gamma)}, \tag{17}$$

where m_j is the mass of an atom of the condensing j-th gas; $\bar{m} = x_j m_j + (1 - x_j) m_g$; x_j is the concentration of the j-th atoms; m_g is the mass of an atom of the gas-diluent. Introducing the dimensionless quantities $\bar{n} = n\sigma^3$, $\bar{r}_* = r/\sigma$, we write the condition dz = const in the form

$$\bar{r}_* \bar{n}_{j0} (\bar{m}/m_j)^{0,5} \bar{T}^{\gamma/2(1-\gamma)} = \text{const}. \tag{18}$$

For a flow of a one-component gas, the condition of constancy of the mean cluster size with a prescribed channel geometry corresponds to points in the plane $\log \bar{n}_0 - \log \bar{T}_0$ lying between lines of equal entropy and dz = const. Considering the relative closeness of these lines, for simplicity we will check the results of the experiment in [20] for satisfaction of dz = const.

The initial data: $T_0 = 295$ K, x = 0.06, d = const ($r_* =$ const). The points of the beginning of condensation: $p_0^{Ar} \approx 6 \cdot 10^5$ Pa, $p_0^{Kr} \approx 4.2 \cdot 10^5$ Pa, $p_0^{Xe} \approx 3.4 \cdot 10^5$ Pa. The trend of the calculations is clear from Table 1. Some of the parameters in the table were calculated to within the constant multiplier. Thus, with allowance for T_0 = const, instead of $\bar{n}_{j0}$ we calculated the values $p_{0j}\sigma^3 \sim \bar{n}_{j0}$. These values were corrected for "one" nozzle geometry. Here, we used the scaling law $\bar{p}_0\bar{d}_0^{0.8}$ = const and we considered the difference in $\bar{m}/m_j$. The result of such a correction and allowance is the quantity $\langle \bar{n}_{j0} \rangle$. Values of the latter are shown in Fig. 2 as a function of $\bar{T}_0$. Also shown there for comparison is the line dz = const at $\bar{r}_*(\bar{m}/m_j)^{0.5}$ = const. It is evident that Ar and Kr are generalized fairly well. The exception of Xe in this regard can be explained on the one hand by the effect of $\bar{m}$ on Dt [see (16)] and, on the other hand, by the fact that this exception is traditionally observed (similar results can be found in [18]) and the inclusion of this element in a single group of similar gases (Ar, Kr) is relative.

Scaling laws for homogeneous condensation in expanding supersonic gas flows are approximate in character and are of limited application. It can be stated that these laws exist due to a very low degree of correlation, and they are a manifestation of the "conservation laws" for the maximum supercooling ϑ_m and the saturation temperature T_s.

However, the laws are in fairly good agreement with the latest empirical findings and will undoubtedly prove useful both for planning gasdynamic experiments on condensation and for correctly understanding the results of such studies.

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